2011年度博士論文要旨

Intermolecular Interaction, Crystallization, and Phase Behavior of Poly(3-hydroxybutyrate) and Cellulose Acetate Butyrate Blends Investigated by Infrared, Near Infrared Spectroscopies and Synchrotron Radiation SAXS/WAXD Techniques

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Recently, one of the serious environmental problems is the “waste” induced by using petroleum-based plastics, which has been resulting in vast loss of the government budget for the management and destruction. The use of biodegradable plastics to replace petroleum-based plastic is one expected way to solve this problem in the world. Poly(3-hydroxybutyrate) (PHB) is the most abundant bacterial polymer with perfect biodegradability, however, it has several weak points for practical industry applications, such as, its rigid and high melting properties and the relatively expensive price. In this thesis, the derivatives of cellulose as Cellulose Acetate Butyrate (CAB) were selected to blend with PHB due to its low price, abundant sources and the wide utilization in industry field for accelerating the application of PHB. For obtaining the fundamental understandings about this blend, its thermal properties, intermolecular interactions, composition fluctuations, crystallization and melting behaviors were mainly explored in this thesis by operating heating and isothermal processes. These researches were operated by using differential scanning calorimetry (DSC), Infrared (IR) spectroscopy, simultaneous synchrotron small-angle X-ray scattering (SAXS), wide angle X-ray diffraction (WAXD) as well as infrared and near-infrared imaging techniques in conjunction with chemometrics, e.g., principle component analysis (PCA) technique. The novelty and originality of this thesis can be described as follows: the first point is that the exchanges between the intra-molecular hydrogen-bonding interactions of the neat PHB (C=O•••H–C) and the intermolecular interactions of CAB and PHB (C=O•••H–O) were interpreted through investigating various ratios of blends with temperature-dependent IR spectroscopy and combining with the results of thermal behavior that obtained from DSC and the crystal structures from WAXD. The second point is that the effect of the intermolecular hydrogen bonding and the effect of molecular weight in the overall crystallization process, from the amorphous melt step to the intermediate state, further

to the primary crystallization process, and the secondary crystallization process, were deeply explored with IR spectroscopy. The third point is that the phase behavior of PHB and CAB in the PHB/CAB blend was explored by using the simultaneous WAXD and SAXS measurements in one heating process. Two glass transition points of the blend were detected and the crystal structures at high temperature were studied. The fourth point is that the conformational evolution and difference of the spherulite during the isothermal crystallization process of the PHB/CAB blend were studied by using FT-NIR imaging spectroscopy. The images of specific bands ratios in the regions of 1^{st} overtone C=O and the 2^{nd} overtone C=O stretching vibration were purposed. Multivariate analysis as PCA was used to analyze the heterogeneity of various localized NIR spectra.